

Application No. 09/764,001

Filed: January 17, 2001

Group Art Unit: 1754

REMARKS

1. This is in response to the Office Action mailed February 27, 2003. Claims 1, 15-17, 19-25, 28 and 29 remain pending in this application.

2. Applicant requests reconsideration of the rejections under 35 USC 103.

a. The present invention relates to a catalyst on a support for the selective oxidation of sulfur-containing compounds. As a catalytically active material, it comprises a mixed oxide having atomically mixed iron ions and zinc ions in an oxidic lattice. The specific surface area of the catalyst is more than 20 m<sup>2</sup>/g and exhibits substantially no reversing Claus activity under the reaction conditions of said oxidation.

The inventors have found that it is possible to provide a catalyst with a high specific surface while having a high selectivity for oxidation of sulfur-containing compounds.

b. Applicant would like to emphasize the differences in reaction mechanisms in a process wherein a catalyst according to the

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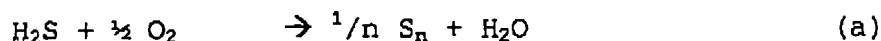
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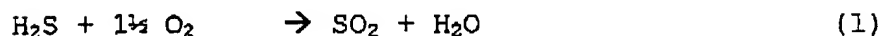
invention may be used and the process described in US-A-5,965,100.

Further detail on this issue follows.

(1) Selective oxidation, catalyzed by a catalyst according to the invention, involves the following reaction:



The activity for the following side-reactions is very low in a catalyst according to the invention



It is an advantage of the selective oxidation catalyst of the invention that it is essentially not affected by the presence of water, because the reaction steps are not equilibrium reactions. It is a further advantage that a high selectivity is achievable at a large stoichiometric excess of oxygen.

(2) US-A-5,965,100 relates to a Claus process wherein use is made of a catalytic system that catalyses a different reaction mechanism. The catalyst is a combination of a conventional Claus

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catalyst and a special catalyst comprising a mixture of metal oxides or a mixed metal oxide. As a carrier, activated alumina is *inter alia* suggested, which is a Claus active support. The specific surface area of the catalyst is at least 6 g/m<sup>2</sup>, but it is nowhere suggested to use a catalyst with a specific surface area of more than 20 g/m<sup>2</sup>.

The catalyst system catalyses the direct oxidation of H<sub>2</sub>S to sulfur. Direct oxidation is different from the selective oxidation reaction, catalyzed by a catalyst according to the present invention. Direct oxidation catalysis can be regarded as a "Claus factory on a molecular scale". H<sub>2</sub>S does not directly react with oxygen to sulfur, but to SO<sub>2</sub> (as a result of the high oxidation activity). Simultaneously, the direct oxidation catalyst is a highly active Claus catalyst, which reacts the formed SO<sub>2</sub> together with unreacted H<sub>2</sub>S to sulfur, in a Claus reaction:

*direct oxidation* Oxidation:  $\text{H}_2\text{S} + 1\frac{1}{2} \text{O}_2 \rightarrow \text{SO}_2 + \text{H}_2\text{O}$  (1)

Claus reaction  $2 \text{H}_2\text{S} + \text{SO}_2 \leftrightarrow \frac{3}{n} \text{S}_n + 2 \text{H}_2\text{O}$  (2)

c. From the office action page 12, first paragraph it appears that the Examiner incorrectly assumes that the catalyst of US-A-5,965,100 may exhibit essentially no reversing Claus activity.

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However, as the Claus reaction is an equilibrium reaction, the direct oxidation catalyst - such as the catalyst of US-A-5,965,100 - by definition catalyses reaction (2) in both directions. Thus, it also catalyses the reversing Claus reaction (reaction from elemental sulfur to  $H_2S$  and  $SO_2$ ), until the equilibrium is reached.

This is also clear from the description of the preferred embodiment. At column 4 of '100, lines 59-65, it is specified that  $H_2S$  is oxidized by a number of major reactions, namely the oxidation of  $H_2S$  to  $SO_2$  (1), and the Claus reaction (2), wherein the arrows indicate that the Claus reaction also takes place in the reverse direction.

As a result of this Claus activity, the sulfur yield is strongly reduced at high water vapor contents in the feed. This makes direct oxidation catalyst unsuitable for feeds with high water contents.

d. Another drawback of this process is the requirement to accurately control the oxygen dosage, in order to maintain the  $H_2S$  to  $SO_2$  ratio at 2 : 1 (for maintaining reaction (2) in the rightward direction).

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Thus, the present catalyst clearly catalyses a different type of reaction mechanism.

e. US-A-5,891,415 relates to a process for the selective oxidation of hydrogen sulfide, wherein use is made of a solid porous catalyst (preferably) consisting essentially of oxides of zinc and iron. The catalyst is preferably not a catalyst on a support (see claim 1 & col 4, lines 35-38). The catalyst has a surface area of about 1-5 m<sup>2</sup>/g (col 4, lines 31-32).

The Examiner states that the difference between Applicant's claim 1 and US-A-5,891,415 is that Applicant's claim 1 sets forth that the iron and zinc ions are present in the form of an atomic mixture in the oxidic lattice, whereas US-A-5,891,415 does not mention that their catalyst is in spinel form.

Further the Examiner acknowledges that the present claim 1 differs from US-A-5,891,415 in that the surface area of the catalyst according to the present invention is more than 20 m<sup>2</sup>/g.

f. Applicant does not believe that it would be obvious to modify the catalyst of US-A-5,891,415 according to the teachings of US-A-5,965,100 for the following reasons.

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(1) First, the present invention and US-A-5,891,415 on the one hand, relate to a selective oxidation process. US-A-5,965,100, on the other hand relates to a direct oxidation process, making use of a catalyst that should catalyze a different type of reaction. In particular the direct oxidation catalyst shows Claus activity and also reversing Claus activity (see above), whereas the catalyst of the present invention substantially does not.

Thus the Examiner is combining a non-preferred catalyst of US-A-5,891,415 (namely a catalyst on a support/carrier; see column 4, lines 34-37) with a specific feature of the catalyst of US-A-5,965,100, which catalyst is intended to catalyze a completely different reaction mechanism. Moreover that specific feature (the specific surface area being more than 20 m<sup>2</sup>/g) is not even explicitly disclosed in US-A-5,965,100.

(2) Applicant does not believe that it would be obvious to modify the catalyst of US-A-5,891,415 by increasing its surface area because of its expected advantage.

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The Examiner should recognize that US-A-5,891,415 relates to the selective oxidation of hydrogen sulfide. Thus the skilled person, being aware of the fact that selectivity in oxidation of hydrogen sulfide is generally thought to be adversely affected by an increase of specific surface area, would conclude that the specific area of 1-5 m<sup>2</sup>/g would be essential to oxidize the hydrogen sulfide with a satisfactory selectivity. He would contemplate that an increased specific surface area would lead to a reduction in selectivity, and consider this as a disadvantage.

A catalyst for selectively oxidizing hydrogen sulfide in principle catalyses the following exothermic two-step reaction:



The reaction is selective for the first step (a). When selectively oxidizing the H<sub>2</sub>S, it is typically desired that the elemental sulfur has moved out of the catalyst particle (wherein it has been formed) before the subsequent reaction step (b) has occurred at a significant rate.

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When increasing the specific area, the reaction rate of both reaction steps increases. As a result, the heat production per second is increased, which in turn leads to an increase in temperature of the catalyst particles. As the temperature rises, the reaction rate of step (b) is expected to increase relatively to the reaction rate of step (a). Thus the selectivity is expected to decrease.

In fact, the reasonable expectation of a loss of selectivity due to a higher specific surface area may also be concluded from US-A-5,965,100. This publication teaches that spinels have a high tendency to form SO<sub>2</sub>. This is illustrated by the reaction scheme at column 6, lines 3-15, which shows that H<sub>2</sub>S is directly converted to SO<sub>2</sub> and at column 4, line 63, see in particular the reaction schemes. From this, the skilled person would also conclude not to use a catalyst with an increased specific surface if he wants to selectively oxidize the H<sub>2</sub>S.

In summary, from the desire to selectively oxidize sulfur-containing compounds wherein formation of SO<sub>2</sub> is largely suppressed or even avoided, the skilled person would conclude from US-A-5,891,415 that a relatively low specific surface area of 1-5 m<sup>2</sup>/g is essential in order to maintain a good selectivity. He



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would not consider increasing the specific area for fear of loosing selectivity. Even from US-A-5,965,100 he would not be motivated to increase the specific surface area, since it does not teach him anything on the behavior of selective oxidation catalysts. In fact he may even expect an (undesired) increase in SO<sub>2</sub> formation.

Thus, it is surprising that a catalyst according to the invention displays satisfactory selective oxidation catalytic activity in combination with a high specific surface. Hence, the present invention is non-obvious over US-A-5,891,415 in view of US-A-5,965,100.

(3) The Examiner further rejects the method claims 21-23 as being unpatentable over US-A-5,891,415 in view of US-A-5,965,100 and further in view of JP 56-10,338.

JP 56-10,338 relates to a method for making a catalyst of the spinel type on a carrier in the presence of Al polychloride.

It is nowhere suggested that chloride plays a specific role in the formation of such a spinel. The use of an aqueous aluminum chloride solution may be because of a strong acidity of the

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solution, which helps to achieve a homogenous impregnation of the carrier. In particular there is no suggestion in this publication to prepare a catalyst as defined in claim 1 specifically of a mixed oxide type of Fe and Zn with a high surface area yet a good selectivity for oxidizing sulfur containing compounds.

In any case, the abstract of JP 56-10,338 does not mention the feature specified in claim 23, namely to use chloride in an amount of 0.1-20 wt. %. There is no suggestion that such an amount of chloride would have an beneficial effect. Thus, the subject-matter of this claim is non-obvious over the cited prior art, irrespective of the non-obviousness of claim 1.

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The Examiner is encouraged to telephone the undersigned attorney to discuss any matter that would expedite allowance of the present application.

Respectfully submitted,

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